

**Comment on “A Humidity-controlled Fast Integrated Mobility Spectrometer (HFIMS) for rapid measurements of particle hygroscopic growth” by Tamara Pinterich et al.**

We thank the reviewers for their constructive comments. Please find below detailed responses to each comment or question, including notations of improvements to the manuscript. Reviewer comments are *italic*. Changes to the text are in **blue font** and underlined.

**Anonymous Referee #1**

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*This work reports on a new way to quantify aerosol hygroscopic growth factors. The authors introduce the Humidity-controlled water-based Fast Integrated Mobility Spectrometer (HFIMS). The topic of this paper certainly fits in this journal and the paper is very well-written and to the point. The new instrument is a good addition and highly relevant for ambient applications that require fast time resolution. The instrument provides some advantages over other recent instrument designs that conduct growth factor measurements. Characterization tests were performed that show that the instrument can accurately quantify growth factors and deliquescence/efflorescence RH values for a known inorganic salt. The authors can strengthen their manuscript by demonstrating that the instrument works well on a mobile platform. If this is not possible for some reason, the authors should mention why this is not possible; from what i can tell, the field data shown were from a stationary set-up.*

Specific Comments:

1. *Figure 2: "Sheat flow" spelled wrong (Sheath)*

We replaced “Sheat” with “Sheath” in Figure 2 and thank the reviewer for pointing out the misspelling.

2. *The authors should provide a clear statement about the range of dry diameters the instrument can handle for the growth factor measurement. is it 35-165 nm as Lines 6-7 suggest on page 6? state it clearly and compare it to other available instruments.*

We thank the reviewer for this suggestion. Under the operation conditions in this study, the HFIMS dry particle size range is 15 to 205 nm. Available systems based on optical or aerodynamic sizing (i.e., DASH-SP or H-DMA-APS) are better suited for measuring the hygroscopicity of larger particles. We note that the size range for DMA or WFIMS depends on sheath flow rate and classifying voltage. For example, the upper particle size range could be increased by operating DMA or WFIMS at a lower sheath flow rate. On the other hand, a lower sheath flow rate leads to either a reduced sampling rate (when the ratio of sheath to aerosol flow rate is maintained) or a reduced size resolution (when the aerosol flow rate is maintained). In this study, the WFIMS was operated to cover the smaller size range, in particular, the 5 standard sizes ranging from 35 to 165 nm suggested by the EUSAAR project. Following the

reviewer’s suggestion, we now clearly stated the HFIMS’ dry particle diameter range, and included the size range of other available instruments for comparison. Following paragraph and table were added to the manuscript (p.5, lines 21 – 26):

“In this study, WFIMS separating voltages ranged from 70 to 4500 V, allowing hygroscopicity measurement for particles with dry diameters ranging from 15 to 205 nm. This includes the standard sizes from 35 to 165 nm suggested by the EUSAAR project (Duplissy et al. 2009). Dry particle size ranges of the HFIMS and other representative instruments are shown in Table 1. Compared to HFIMS, systems based on optical or aerodynamic sizing have a larger upper size limit. On the other hand, HFIMS is capable of rapid measurement of particles with dry diameters below 150 nm, which are difficult to detect using an OPC or APS.”

Table 1 Dry particle diameter range ( $D_{p,0}^{\min}$  -  $D_{p,0}^{\max}$ ) of four particle hygroscopicity instruments including HFIMS. Diameter ranges of HFIMS, BMI HTDMA and DASH-SP were estimated based on growth factors ranging from 0.8 to 2.2. DASH-SP dry size range was estimated for 5 l/min DMA sheath flow rate, 281 V minimum classifying voltage and 135 nm cut size of the OPC as specified in Sorooshian et al. (2008). The particle dry diameter range of BMI HTDMA was calculated for a typical fieldwork configuration (i. e.  $Q_a=0.6$  l/min,  $Q_{sh}=6$  l/min, Lopez-Yglesias et al., 2014) and classifying/scanning voltages ranging from 20 V to 6000 V. The diameter range of H-DMA-APS is listed as specified in Leinert & Wiedensohler (2008).

<u>Instrument</u>	<u><math>D_{p,0}^{\min}</math> [nm]</u>	<u><math>D_{p,0}^{\max}</math> [nm]</u>
<u>HFIMS (This study)</u>	<u>15</u>	<u>205</u>
<u>BMI HTDMA (Lopez-Yglesias et al. 2014)</u>	<u>33</u>	<u>425</u>
<u>DASH-SP (Sorooshian et al. 2008)</u>	<u>170</u>	<u>455</u>
<u>H-DMA-APS (Leinert &amp; Wiedensohler 2008)</u>	<u>800</u>	<u>1600</u>

We also highlighted the advantage of DMA-OPC and DMA-APS systems for measurements of larger particles (p. 2 lines 23 – 25):

“These systems, based on optical sizing or aerodynamic sizing, are capable of measuring the hygroscopicity of large accumulation mode particles, which are important to evaluating the optical properties and direct radiative effects of ambient aerosols.”

- It is nice that it can detect smaller sizes where there may be higher concentrations, but it may be useful to point out that larger sizes may be more relevant for light-scattering and radiative forcing? For what purposes are smaller sizes most relevant (e.g., health effects)?*

We thank the reviewer for the suggestion and comment. The manuscript was revised accordingly:

P. 2, lines 23 – 25:

[“These systems, based on optical sizing or aerodynamic sizing, are capable of measuring the hygroscopicity of large accumulation mode particles, which are important to evaluating the optical properties and direct radiative effects of ambient aerosols.”](#)

P. 2, lines 32-34 and p. 3, lines 1 – 2:

[Compared to systems based on optical or aerodynamic sizing, HFIMS extends fast measurements to particles with diameters below 150 nm. Particles smaller than 150 nm often represent a large fraction of cloud condensation nuclei population, and may have a strong impact on human health \(Chen et al. 2016\).”](#)

4. *A few times it is stated that "...size distribution spanning a factor of ~3 in particle diameter". Rather than using the confusing "factor of ~3" everywhere, can the authors simply just state the range of diameters?*

The size range of humidified particles that can be measured simultaneously by WFIMS depends on the separating voltage. At a constant separating voltage WFIMS can measure size distributions spanning a factor of about 3 in particle diameter (a factor of 10 in electrical mobility) simultaneously. The actual diameter range of the WFIMS is therefore varied according to the diameter of dry particles classified by the DMA.

For example, at a particle dry diameter of 35 nm WFIMS separating voltage is set such that humidified particle with diameters ranging from 28 nm (GF = 0.8) to 95 nm (GF = 2.7) can be measured simultaneously. HFIMS growth factor ranges for all particle dry sizes investigated in this study are given in SI section S4.

5. *What is the residence time of aerosol in different parts of the instrument?*

Long DMA ( $Q_{sh} = 3$  l/min,  $Q_a = 0.3$  l/min)  $\rightarrow 7.4$  s

Nano DMA ( $Q_{sh} \rightarrow 3$  l/min,  $Q_a = 0.3$  l/min)  $\rightarrow 0.8$  s

RH control unit and tubing between DMA outlet and WFIMS inlet ( $Q_a = 0.3$  l/min)  $\rightarrow 6.5$  s

WFIMS  $\rightarrow 1.5$  s – 2.6 s depending on particle trajectory

The particle residence time inside the WFIMS depends on particle trajectory. The residence time for each measured particle can be calculated from its detected position and explicitly accounted for (Olfert et al., 2008). We also note that in this initial study, the residence times inside the DMA and between DMA outlet and WFIMS inlet were not optimized. For example, the residence time inside the long DMA could be reduced by operating the DMA at sheath and aerosol flow rates of 10 l/min and 1 l/min, respectively, without negative impact on dry particle size range or sampling rate.

6. *How long does it take to scan through a range of different relative humidities? Provide numbers and compare to other instruments.*

Hygroscopicity of ambient particles is typically measured by HTDMA at a single RH for a range of particle sizes, similar to results shown in Figure 10. We agree that measurement at multiple RH provides a more complete characterization of particle hygroscopicity. The current RH control unit was not optimized for stepping the RH of HFIMS, such that it takes several minutes for the RH to stabilize at the new setpoints. Optimization of the RH control unit, possibly using a similar design as reported in Lopez – Yglesias et al. (2014) will be included in future improvements.

7. *What is the range of relative humidities the instrument can handle?*

Currently stable RH conditions can be maintained from 20% - 90% as shown in Figure 5.

8. *Section 2.1: Can the authors comment on hysteresis effects that are common with nafion based systems?*

The RH of aerosol sample exiting the Nafion exchanger is measured by an RH sensor immediately down-stream of the exchanger, and the RH measurement is input to a PID module, which controls the aerosol sample RH (Fig. 1). Therefore, we expect the potential hysteresis effects of the Nafion exchanger has negligible impact on aerosol sample RH.

9. *Has the instrument been deployed on a mobile platform? The paper states that the instrument is ideal for mobile platform work but it is unfortunate that data does not appear to be shown from its use in the field to give readers a better sense of how robust it is when the conditions aren't 'easy' such as in a lab or at a stationary field site.*

The instrument has not (yet) been deployed on a mobile platform. We removed the part ‘... or measurements onboard mobile platforms.’ from the last sentence of the conclusions section (p. 9, lines 24 – 25).

10. *Figure S2: x-axis has "distance" misspelled.*

We replaced “Distande” with “Distance” in Figures S2 (a) and (b) and thank the reviewer for pointing out the misspelling.

**Anonymous Referee #2**

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*A new instrument for fast measurement of particle hygroscopicity, HFIMS, is reported in this paper. Different from the existing fast hygroscopicity measurement instruments, HFIMS measures the electrical mobility diameters of the grown particles so that particle density and refractive index are not needed anymore. Compared with the HTDMA, HFIMS avoids the diameter scan which is time consuming therefore has a much higher time resolution and may work on mobile platforms. The topic fits well in AMT and the manuscript is well written. I therefore recommend the final publication of this paper on AMT.*

Specific comments:

- 1. What is the maximum RH the system can reach? How about its stability at different RH levels? Since the RH is only measured before the flow entering WFIMS (not like HTDMA in which RH is also measured at the sheath exit), the real RH the particles undergo may be slightly different from the value given by RH sensors (e.g. in case of high particle concentration and very slow growth, or temperature difference between parts in the system). Does the author have any comment on this?*

With the current RH control unit the maximum RH that the system can hold stable is 90%. As pointed out by the reviewer it might be possible, that the RH during mobility classification slightly differs from the measured sheath/aerosol RH. However, data presented in Fig. 5 suggest that these differences are minor. Thermal insulation of the RH control section and WFIMS separator could further reduce temperature differences between relevant parts, and will be part of future improvements.

To answer the reviewer's comment on the effect of water vapor depletion, we estimated the change in RH due to water uptake at standard temperature and pressure for a growth factor of 2 and dry particle sizes up to 200 nm. For DMA classified ambient particles with concentrations up to  $10^4 \text{ cm}^{-3}$  the change in RH (initially 85%) due to water vapor uptake inside the WFIMS separator should be less than 0.1%.

- 2. What is the time needed to washout all particles in the system at a typical setting of flow rates? The washout time may strongly influence the time resolution of the measurement.*

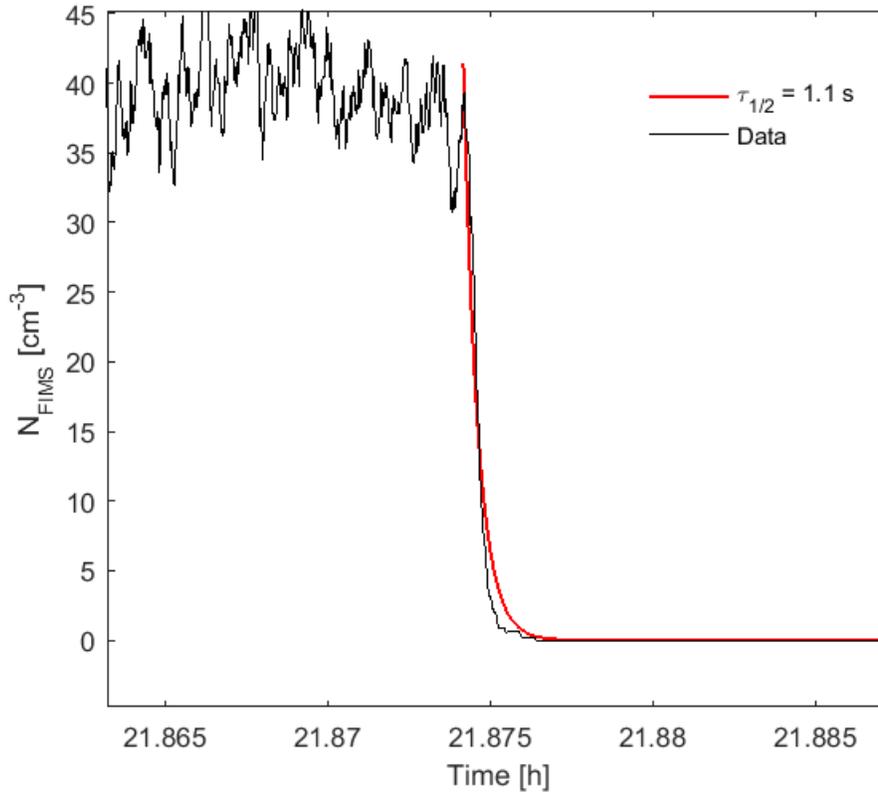


Figure R 1 Time evolution of particle number concentration  $N_{FIMS}$  measured by WFIMS (black line) showing a steep concentration drop after setting nano DMA voltage to 0 V ( $t \sim 21.875$  h). Data around the concentration drop were fitted using Eq. (R1) to determine the half-life  $\tau_{1/2}$  for concentration decay (Eq. R2).

$$N_{FIMS}(t) = N_0 \cdot e^{-\lambda \cdot t} \quad (\text{R0})$$

$$\tau_{1/2} = \frac{\ln(2)}{\lambda} \quad (\text{R0})$$

In this study the washout time was determined from the response of FIMS after setting the nano DMA voltage to 0 V, which creates a nearly stepwise change of classified particle concentration down to  $0 \text{ cm}^{-3}$ . Particle concentrations measured by WFIMS, i.e.  $N_{FIMS}$  (black line in Fig. R1), were fitted (red line in Fig. R1) around the steep drop ( $t \sim 21.875$  h) using an exponential decay model (Eq. R1). The half-life  $\tau_{1/2}$  (Eq. R2) of particles within the system was found to be 1.1 s. In other words it takes the HFIMS about 4.4 seconds ( $4 \cdot \tau_{1/2}$ ) to washout 94% of all particles.

3. *The finite-width transfer function of WFIMS may cause a smooth effect in the measured distribution of GF. The discussion about it (P7L10) is a bit too brief. Can the author give a more detail discussion on the uncertain of GF distribution due to this effect? Is it possible and necessary to correct this smoothing effect?*

As the reviewer pointed out correctly, the measurement signal of the HFIMS is a smoothed integral transform of the particle's actual growth factor probability distribution function (GF-PDF) together with the transfer functions of the DMA and WFIMS. Hence, a data inversion algorithm should be applied to the response signal of the HFIMS measurements to retrieve the GF-PDF. Because DMA transfer function, WFIMS transfer function, and the response of the HFIMS are known (Stolzenburg & McMurry, 2008; Kulkarni and Wang, 2006), the actual GF-PDF could be solved by the data inversion using an algorithm similar to TDMAfit (Stolzenburg & McMurry, 1988, 2008) and TDMAinv (Gysel et al., 2009). The development and demonstration of this algorithm are part of an ongoing study.

#### References:

- Gysel, M., McFiggans, G., & Coe, H. (2009). Inversion of tandem differential mobility analyzer (TDMA) measurements. *Journal of Aerosol Science*, 40(2), 134-151.
- Kulkarni, P., & Wang, J. (2006). New fast integrated mobility spectrometer for real-time measurement of aerosol size distribution—I: Concept and theory. *Journal of Aerosol Science*, 37(10), 1303-1325.
- Stolzenburg, M., & McMurry, P. (1988). TDMAFIT user's manual. University of Minnesota, Department of Mechanical Engineering, Particle Technology Laboratory, Minneapolis, 1-61.
- Stolzenburg, M., & McMurry, P. (2008). Equations governing single and tandem DMA configurations and a new lognormal approximation to the transfer function. *Aerosol Science and Technology*, 42(6), 421-432.

4. *Fig. 5: It seems that the blue dots are on the extension of the deliquescence part of the solid line (reported by Cruz and Pandis, 2000) but the red dots are a bit lower. Does the author have any explanation on this difference? Is it possible to add error bars as measurement uncertainty?*

We agree with the reviewer that the efflorescence data (blue dots) appear to agree better with data reported by Cruz and Pandis (2000) than the deliquescence data (red dots). Overall, the differences between our deliquescence data and results reported by Cruz and Pandis (2000) are quite minor. Vaisala humidity sensors (HMP60) used in this study have a typical accuracy of  $\pm 3\%$ . The differences between deliquescence data may be due to the uncertainty in RH measurements. We added corresponding error bars to measured data in Fig. 5, and revised the figure caption. Implementation of more accurate RH measurement is planned for future improvement.

5. *P5L6: It will be clearer if the definition of mobility resolution  $R$  is also given here.*

As suggested by the reviewer we adapted the sentence (p. 5, lines 10 – 11) as follows:

“Knowing  $Z_{p,\max}^*$  the mobility resolution  $R$ , [which is defined as the ratio of particle mobility  \$Z\_p\$  to the full width at half height of the WFIMS transfer function  \$\Delta Z\_p^\*\$](#) , can be calculated according to (Kulkarni and Wang, 2006a) ...”

And added the term  $\underline{z}_p/\Delta z_p^*$  to Eq. (3)

6. *Fig. 7: What is growth factor resolution, the ratio between growth factor and its uncertainty? It is better to give a clear definition in the text.*

We thank the reviewer for pointing out this inconsistency. Figure 7 actually shows the mobility resolution, i. e. particle mobility to the full width at half height of mobility transfer function, over the growth factor range of 1-2.2. Changes were made to both the label of Fig. 7 and the reference to Fig. 7 in the text (p. 7, line 25).

# A Humidity-controlled Fast Integrated Mobility Spectrometer (HFIMS) for rapid measurements of particle hygroscopic growth

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**Abstract.** We present a Humidity-controlled Fast Integrated Mobility Spectrometer (HFIMS) for rapid particle hygroscopicity measurements. The HFIMS consists of a differential mobility analyzer (DMA), a relative humidity (RH) control unit and a water-based FIMS (WFIMS) coupled in series. The WFIMS (Pinterich et al., 2017) combines the Fast Integrated Mobility Spectrometer (Kulkarni and Wang, 2006a, b) with laminar flow water condensation methodologies (Hering and Stolzenburg, 2005; Spielman et al., 2017). Inside the WFIMS, particles of different electrical mobilities are spatially separated in an electric field, condensationally enlarged and imaged to provide 1-Hz measurements of size distribution spanning a factor of ~3 in particle diameter, sufficient to cover the entire range of growth factor for atmospheric aerosol particles at 90 % RH. By replacing the second DMA of a traditional hygroscopicity tandem DMA (HTDMA) system with the WFIMS, the HFIMS greatly increases the speed of particle growth factor measurement.

The performance of the HFIMS was evaluated using NaCl particles with well-known hygroscopic growth behavior, and further through measurements of ambient aerosols. Results show that HFIMS can reproduce, within 2 % the literature values for hygroscopic growth of NaCl particles. NaCl deliquescence was observed between 76 % and 77 % RH in agreement with the theoretical value of 76.5 % (Ming and Russell, 2001), and efflorescence relative humidity (43 %) was found to lie within the RH range of 41 % to 56 % reported in the literature. Ambient data indicate that HFIMS can measure the hygroscopic growth of five standard dry particle sizes ranging from 35 to 165 nm within less than three minutes, which makes it about an order of magnitude faster than traditional HTDMA systems.

## 1 Introduction

The hygroscopicity of atmospheric aerosols is a key parameter in determining their impact on global climate. The uptake of water by individual particles increases the light scattering, enhances heterogeneous chemical transformations important to secondary aerosol formation (e.g., Surratt et al., 2010), and is important in the formation of cloud droplets. The abundance of hygroscopic particles that act as cloud condensation nuclei affects cloud formation and cloud droplet number concentrations, which in turn influences cloud albedo, coverage and lifetime ([Twomey 1977; Albrecht 1989](#)). These “indirect effects” of atmospheric aerosols on the Earth’s radiation balance remain one of the largest uncertainties in understanding climate change

(IPCC 2013). Hygroscopicity is among the key determinants of the ability of aerosol particles to form cloud droplets and therefore the aerosol indirect effects (e.g. Mei et al., 2013; Liu and Wang, 2010).

Most commonly particle hygroscopic growth is measured using hygroscopicity tandem differential mobility analyzer (HTDMA) systems, which consist of two differential mobility analyzers (DMAs) in series, separated by a means to control the sample flow relative humidity (RH). HTDMA systems first select a single particle size using the first DMA, change its relative humidity environment, then scan the classifying voltage of the second DMA to measure the distribution of particle sizes resulted from the change in RH. The HTDMA method is accurate, but slow. Typically the time required to complete a measurement cycle of determining the growth factor at a single relative humidity (such as 90 %) for 5 different particle sizes is about 30 min (e. g. Cerully et al., 2011). Measurement periods are especially long for large particles which are low in concentration, and small particles that have a low charging efficiency.

Several investigators have worked to increase the speed of HTDMA measurements by replacing the second DMA with an instrument that is capable of fast size distribution measurements. Sorooshian et al. (2008) developed a Differential Aerosol Sizing and Hygroscopicity Spectrometer Probe (DASH-SP), in which wet particle size is measured by an optical particle counter (OPC). By replacing the 2nd DMA with an optical counter, DASH-SP accelerates the measurement significantly. However, the optical counting limits DASH-SP measurements to particles larger than ~150 nm in diameter, and a sophisticated algorithm is required to account for the variation of particle refractive index due to water uptake and its impact on optical sizing. Stolzenburg et. al (1998) developed a high-flow mobility analyzer which they coupled to an optical particle counter, with humidity control upstream of the mobility size separation, and an aerosol dryer downstream to measure particle size change upon dehumidification. Leinert and Wiedensohler (2008) developed a DMA-aerodynamic particle sizing (APS) system to examine growth factors, but their measurements were complicated by the change in particle density, which affects the aerodynamic measurement. While faster, the limitation of these coupled DMA-optical counter or DMA-aerodynamic sizing techniques is the limitation on the measurement size range and the additional complexity or uncertainty in mapping the optical or aerodynamic size onto the physical size of the particle that is important to assessing water uptake. [These systems, based on optical sizing or aerodynamic sizing, are capable of measuring the hygroscopicity of large accumulation mode particles, which are important to evaluating the optical properties and direct radiative effects of ambient aerosols.](#)

To address the need for fast and precise measurements of particle hygroscopic growth, we have developed a Humidity-controlled water-based Fast Integrated Mobility Spectrometer (HFIMS), which replaces the second DMA of the HTDMA systems with a water-based FIMS (WFIMS; Pinterich et al., 2017). By detecting particles of different sizes simultaneously, WFIMS provides rapid measurements of the size distribution of humidified particles. Unlike the final optical sizing of Sorooshian et al. (2008), or final aerodynamic sizing of Leinert and Wiedensohler (2008), WFIMS measures particle sizes based on electrical mobility. This removes the uncertainty introduced by particle refractive index or density, and provides the same, precise growth factor measurements of the HTDMA systems, but with a much faster measurement speed. ~~Importantly, the HFIMS allows hygroscopicity measurements in the critical size range below 100 nm, which generally dominates atmospheric particle number concentrations.~~ [Compared to systems based on optical or aerodynamic sizing, HFIMS extends](#)

[fast measurements to particles with diameters below 150 nm. Particles smaller than 150 nm often represent a large fraction of cloud condensation nuclei population, and may have a strong impact on human health \(Chen et al. 2016\).](#) Using both laboratory and ambient measurements, we demonstrated that the HFIMS can provide growth factor measurements with 1 % precision for five representative particle diameters in less than 3 minutes - about one order of magnitude faster than traditional HTDMA systems.

## 2 Instrument design

The HFIMS consists of three individual units (see Fig. 1): a TSI DMA (either long-column or nano-column, depending on the particle size) classifying particles at a desired dry size under a low RH, an RH control unit providing independent controls of the size-selected particle sample and WFIMS sheath flow RH, and a WFIMS measuring size distributions of particles after being exposed to a different RH. The WFIMS used here is identical to the original WFIMS (Pinterich et al., 2017), except the HV electrode is replaced with one that provides a uniform electrical field with a small offset from aerosol inlet slit, as described below. This modification is made to optimize the measurements of humidified particle size distributions. In essence, the WFIMS deployed in this study is similar to the alcohol based FIMS reported in Kulkarni and Wang (2006a), except that particle growth is achieved by condensation of water instead of butanol, which is key to hygroscopicity measurements.

### 2.1 Relative humidity control (RH-Control)

An automated RH control system was constructed to independently control the RH of the size selected aerosol sample flow ( $RH_a$ ), and the WFIMS sheath flow ( $RH_{sh}$ ). Humid air (>95 % RH) is created by bubbling dry air through sintered metal mufflers submerged in about 20 cm of water. The humid air is then mixed with dry air to provide WFIMS sheath flow. The RH of sheath flow is controlled by a PID controller that drives a proportional solenoid valve on the dry air line based on the sheath flow RH probe reading. The aerosol sample flow RH is controlled using a Nafion® exchanger, and the dry-humid mixture used as the purge flow of this exchanger is controlled independently. As with the sheath flow, the purge flow is obtained by mixing of dry air with that from the humid air source, with a second PID controller that reads the aerosol RH and drives the valve on the dry air line to meet the target aerosol sample RH. To compensate for the evaporative cooling, the bubbler is equipped with a heater and a simple thermostat, set to the room temperature (not shown in Fig. 1).

### 2.2 Configuration of the WFIMS

The WFIMS is identical to the original version presented in Pinterich et al. (2017) except that the HV electrode is replaced with one that provides a uniform electric field with a slight offset from aerosol inlet slit. The WFIMS consists of a parallel plate mobility separator followed by a three-stage condensational growth channel and an imaging system. A particle free sheath flow  $Q_{sh}$  enters at the top, and an aerosol flow  $Q_a$  is introduced through a slit along the entire width of the separator

channel. A constant total flow  $Q_{\text{tot}}$  of 16.5 l/min through the channel is achieved by a vacuum pump along with a critical orifice (O'Keefe Controls Co., No. 55). The desired  $Q_a$  of 0.3 l/min is achieved via PID control of  $Q_{\text{sh}}$  using a flow control valve (MKS, 0248A). The key physical dimensions and operating conditions of the WFIMS in this study are listed in the supplement Sect. S1.

- 5 The WFIMS is configured with a single voltage electrode that has an offset in the direction of the flow ( $z$ -direction). A side-view ( $(x,z)$  - plane) of the WFIMS separator is shown in Fig. 2, where the flow is downward, and the high voltage electrode on the right extending in  $y$ -direction. For measurements of particle growth factor (GF), i.e. the ratio of humidified to dry particle diameter  $D_p/D_{p,0}$ , the size range of the WFIMS only needs to cover the possible change in particle diameter. For ambient particles, the growth factor reported in the literature ranges from 0.8 to 2.2 for RH values up to 90 %. This range  
10 covers the GF of sea salt like particles (Ming and Russell, 2001), and also encompasses the 15 % shrinkage observed for highly agglomerated particles that occurs when the branched structure collapses following the capillary condensation of water (Weingartner et al., 1995). Conveniently, WFIMS operated with a single-voltage electrode has an electrical mobility range of a factor of 10 (Kulkarni and Wang, 2006a, b), which corresponds to more than a factor of 3 in particle diameter, sufficient to cover the GF range of atmospheric aerosols.
- 15 The single high voltage electrode was configured with an offset such that the high voltage region (red area in Fig. 2) begins slightly downstream (32 mm) of the introduction of aerosol into the separator. The offset allows the sheath flow to provide the additional RH conditioning of the size selected sample flow prior to mobility classification as the aerosol flow is less than 2 % of the total flow. As the control of  $\text{RH}_a$  is achieved by using a Nafion exchanger and has a longer response time than that of WFIMS sheath flow, this feature could accelerate the growth factor measurements under different RH by  
20 reducing the waiting time following the change of RH setpoints. However, in the present study we mainly focus on measurements at a single RH (85 %) where this feature is not relevant.

A heater and thermistor were attached near the bottom of the separator to compensate the heat loss to the adjacent cooled conditioner stage (see details in the next paragraph) and to avoid a corresponding change in RH due to this gradient. The heater was driven to equalize the temperatures within the separator. Without heating, the temperature at the bottom of the  
25 separator is about 1.0 °C below that at the top.

Upon exiting the separator the particles continue along their flow trajectories through the three stage growth channel, consisting of conditioner, initiator and moderator, all with wetted walls (Spielman et al., 2017). Particles are enlarged through water condensation without being diverted from their trajectories. WFIMS' three-stage growth channel design provides supersaturation levels of  $\sim 1.35$  across its viewing window, sufficient to activate and grow particles as small as 7 nm  
30 to detectable sizes (Pinterich et al., 2017), while also removing excess water vapor that might otherwise condense on the optical components (Hering et al., 2014). Within the final section, grown particles are illuminated by a laser beam and imaged by a camera at a frame rate of 10 Hz. MATLAB's "Image processing toolbox" is used to detect each droplet and its position. Only particles detected in the center of the channel cross section ( $(x,y)$  - plane) are used for measurements of particle size and concentration in order to avoid the edge effects of electric and flow fields (Olfert et al., 2008). For WFIMS

operated with a single voltage electrode, particle positions can be converted to instrument response mobilities  $Z_p^*$ , using Eq. (22) from Kulkarni and Wang (2006a)

$$Z_p^* = \frac{2(1+\beta)(3\tilde{x}^2 - 2\tilde{x}^3) - \beta}{2+\beta} \cdot Z_{p,\max}^* \quad (1)$$

with flow ratio  $\beta = Q_a/Q_{sh}$  and  $\tilde{x}$  being the ratio of the  $x$ -coordinate of the detected particle to separator gap width  $a$ . Note the conversion of particle position into instrument response mobility is independent of the applied particle enlargement technique (alcohol vs. water). The maximum response mobility  $Z_{p,\max}^*(\tilde{x} = 1)$  measured by the (W)FIMS can be expressed as (Kulkarni and Wang 2006a):

$$Z_{p,\max}^* = \frac{a \cdot Q_a}{\beta(b \cdot l_s \cdot V)} \quad (2)$$

with  $b$  and  $l_s$  being electrode width and length, respectively.  $V$  is the voltage applied across the high voltage electrode.

10 Knowing  $Z_{p,\max}^*$  the mobility resolution  $R$ , which is defined as the ratio of particle mobility  $Z_p$  to the full width at half height of the WFIMS transfer function  $\Delta Z_p^*$ , can be calculated according to (Kulkarni and Wang, 2006a)

$$R = \frac{Z_p}{\Delta Z_p^*} \cong \frac{Z_p}{\beta \cdot Z_{p,\max}^*} \quad (3)$$

Similar to a DMA, the mobility resolution of the (W)FIMS depends on the ratio of sheath to aerosol flow. In addition, (W)FIMS resolution is also a function of particle mobility  $Z_p$ . In DMA classifiers all selected particles traverse the entire mobility separation channel. In contrast, in (W)FIMS systems, only the most mobile particles traverse the entire channel, while the less mobile particles traverse just a portion of the sheath flow. Thus in the (W)FIMS, the mobility resolution reaches the maximum value of  $1/\beta$  only for largest mobility measured (i.e.,  $Z_{p,\max}^*$ ) while the resolution for less mobile particles is lower. When operated at a fixed sheath flow of 16.2 l/min, and an aerosol flow of 0.3 l/min, as is our standard operating configuration, the resolution for those particles that traverse the entire channel is 54, while that for particles traversing just one-half of the channel will be 27.

25 In this study, WFIMS separating voltages ranged from 70 to 4500 V, allowing hygroscopicity measurement for particles with dry diameters ranging from 15 to 205 nm. This includes the standard sizes from 35 to 165 nm suggested by the EUSAAR project (Duplissy et al. 2009). Dry particle size ranges of the HFIMS and other representative instruments are shown in Table 1. Compared to HFIMS, systems based on optical or aerodynamic sizing have a larger upper size limit. On the other hand, HFIMS is capable of rapid measurement of particles with dry diameters below 150 nm, which are difficult to detect using OPC or APS.

### 3. Experimental setup

The capability of HFIMS to accurately characterize particle hygroscopicity is examined by measuring sodium chloride particles, for which hygroscopic growth has been well characterized in prior studies. The experimental setup is shown in Fig. 3. NaCl particles were generated by atomizing a dilute NaCl solution (1.7 mM) using a constant output atomizer (TSI Inc.,

Model 3076), followed by a diffusion dryer. A stable particle number concentration of about  $50 \text{ cm}^{-3}$  was achieved by adapting (i) the diameter of a limiting orifice in dilution stage 1 and (ii) the dry, particle free dilution flow via a needle valve (dilution stage 2). After dilution polydisperse particles were charge equilibrated using a  $^{85}\text{Kr}$  bipolar aerosol neutralizer (TSI Inc., Model 3077) and size selected at low humidity ( $\text{RH} < 10 \%$ ) using a nano-column DMA (TSI Inc., Model 3085). In the present study we selected 50 nm particles. The ratio of DMA aerosol to sheath and sample to excess flow was kept constant at 1:10. During laboratory characterization a CPC (TSI Inc., Model 3010) was operated downstream of HFIMS' DMA in parallel to the RH-Control – WFIMS branch, and it provided the concentration of size selected dry particles. WFIMS was operated at constant separating voltage  $V$  of 1000 V corresponding to a growth factor window of 0.9 to 3.2. A detailed list of HFIMS configuration parameters during the laboratory characterization can be found in the supplement Sect. S2. Note due to non-idealities of the electric field, instrument response mobilities  $Z_p^*$  were calculated using Eq. (1) with an effective voltage  $V_{\text{eff}}$  instead of the applied voltage  $V$  (Eq. (2)). The  $V_{\text{eff}}$  was derived following a calibration procedure detailed in the supplement Sect. S3.

For the measurements of deliquescence and hygroscopic growth of NaCl particles, we matched the relative humidity of aerosol and sheath flows ( $\text{RH}_a = \text{RH}_{\text{sh}}$ ). The measurement of efflorescence was carried out by keeping  $\text{RH}_a$  at 85 % while varying  $\text{RH}_{\text{sh}}$  between 18.8 % and 79.9 %.

The measurement speed of HFIMS was evaluated by sampling ambient aerosols outside of our laboratory at Brookhaven National Laboratory (Upton, New York). Figure 4 shows the schematic of the experimental setup. We obtained ambient particle growth factors (GF) at  $\text{RH}_a = \text{RH}_{\text{sh}} = 85 \%$ , for various dry particle diameters including five diameters (i.e., 35, 50, 70, 110 and 165 nm) used by EUSAAR (European Supersites for Atmospheric Aerosol Research; Duplissy et al., 2009). A detailed list of HFIMS configuration parameters during ambient tests can be found in the supplement Sect. S4. Ambient particles with dry diameter up to 110 nm were classified with a nano-column DMA (TSI Inc., Model 3085), above 110 nm a long-column DMA (TSI Inc., Model 3081) was used. Whereas different DMAs were used to classify particles with different diameters in these preliminary tests, future systems will consist of a single DMA. As a result, the time required to switch between nano- and long-column DMA was not considered when evaluating HFIMS' measurement speed. As the classified dry particle diameter varied, WFIMS separating voltage  $V$  and hence mobility diameter window were adapted accordingly, and the values are listed in the supplement Sect. S4.

## 4. Results and discussion

### 4.1 Laboratory evaluation with NaCl particles

Growth factors of 50 nm NaCl particles measured under both increasing (i.e., deliquescence branch, red diamonds) and decreasing (efflorescence branch, blue circles) RH conditions are shown in Fig. 5. Measurements of deliquescence branch were carried out with matching aerosol and sheath RH (i.e.,  $\text{RH} = \text{RH}_a = \text{RH}_{\text{sh}}$ ). To account for the cubic shape of dry NaCl

particles we used a shape correction factor of 1.08 (Zelenyuk et al., 2006). Hence measured mobility equivalent diameters were decreased by about 4 % to obtain volume equivalent diameters.

The NaCl deliquescence transition observed by HFIMS is just over 76 %, in agreement with the theoretical value of 76.5 % (Ming and Russell, 2001), and measurements by Hämeri et al. (2001), and Cruz and Pandis (2000) of 76 % and 75.6 %, respectively. It should be mentioned that around the deliquescence transition two distinct size modes are observed (see Fig. 6). This suggests some heterogeneity in the RH of aerosol sample (i.e., some particles experienced slightly higher RH than others), which is likely due to temperature variations among different parts of the system. Deliquescence transition data shown in Fig. 5 represent the number weighted mean growth factor for the two modes. Improved RH and temperature control could minimize the RH heterogeneity, and will be a topic of future study. Above the deliquescence transition, growth factors measured by the HFIMS are within 2 % of theoretical values, suggesting the RH heterogeneity has negligible impact on measured particle growth factors above deliquescence RH (e. g. at 85 %).

Figure 5 also shows the efflorescence curve (blue circles), that is the size change when the relative humidity environment decreases. Data were obtained by maintaining  $RH_a$  constant at 85 % while varying  $RH_{sh}$  between 18.8% and 79.9 %. Hence the sheath flow was used to condition the sample RH, a feature made possible by the new offset electrode (see Sect. 2.2). As demonstrated in the supplement Sect. S5 the RH of the mixed flow reaches the average value very quickly, such that the RH becomes uniform at the start of the electric field when the offset electrode is used. Humidified particle diameters  $D_p$  were measured at RH given by:

$$RH = (Q_a \cdot RH_a + Q_{sh} \cdot RH_{sh}) / (Q_a + Q_{sh}). \quad (4)$$

It is common for salt aerosols to exhibit this type of hysteresis, with the droplet becoming supersaturated while gradually decreasing in size until finally recrystallizing at a much lower relative humidity. We observed recrystallization near 43 % RH, which is close to the value of 44 % reported by Biskos et al. (2006), and within the range of 41 % to 56 % (yellow area) reported in the literature.

Using the HFIMS operating conditions listed in the supplement (Sect. S2) we calculated the resolution of its sizing unit, i.e. the WFIMS, as a function of hygroscopic growth factor for non-diffusing particles and compared it to the typical resolution of a DMA, i.e. 10. As shown in Fig. 7 the **mobility GF** resolution of HFIMS is equal to, or exceeds that of the HTDMA over the measured growth factor range, i. e. 1 – 2.27, shown in Fig. 5.

## 4.2 Ambient particle hygroscopicity measurement

Characterization of ambient aerosol hygroscopicity often requires measurements at multiple particle sizes within a reasonable time. This is often challenging for measurements using traditional TDMA systems, especially for the larger particles which are low in number concentration, and the smallest particles which have low charging efficiency.

Figure 8 shows results from ambient measurements with HFIMS, where we evaluated the relative standard error of the mean growth factor (SEM of  $\overline{GF}$ ) as a function of particle counts  $C$ , and corresponding sample duration. Note as the mean particle

growth factor is given by the ratio of wet to dry particle diameter,  $\overline{D}_p$  and  $D_{p,0}$ , respectively, the relative precision in measured  $\overline{GF}$  can be approximated by the relative precision in measured  $\overline{D}_p$  since

$$\frac{SEM(\overline{GF})}{\overline{GF}} = \sqrt{\left(\frac{\partial \overline{GF}}{\partial \overline{D}_p}\right)^2 \cdot SEM^2(\overline{D}_p) + \left(\frac{\partial \overline{GF}}{\partial D_{p,0}}\right)^2 \cdot (\Delta D_{p,0})^2} / \overline{GF} \approx \frac{SEM(\overline{D}_p)}{\overline{D}_p}. \quad (5)$$

Total particle concentrations were relatively low, around  $2200 \text{ cm}^{-3}$ , similar to what might be expected for continental background. The standard error of the mean (SEM) for  $\overline{D}_p$  is estimated as:

$$SEM(\overline{D}_p) = \frac{SD}{\sqrt{C}} \quad (6)$$

where  $C$  is the number of particles counted and  $SD$  the sample standard deviation given by

$$SD = \sqrt{\frac{\sum_{i=1}^C (D_{p,i} - \overline{D}_p)^2}{C}} \quad (7)$$

with  $D_{p,i}$  being the diameter of the  $i$ -th particle measured. The relative SEM of  $\overline{GF}$  can be derived by combining Eqs. (5) and (6):

$$\frac{SEM(\overline{GF})}{\overline{GF}} = \frac{SD}{\overline{D}_p \sqrt{C}}. \quad (8)$$

Note while the sample standard deviation  $SD$ , which is a measure of the dispersion of measured particle sizes from the sample mean diameter  $\overline{D}_p$ , asymptotically reaches the population standard deviation as  $C$  increases (see Figure 8c),  $SEM$ , which is a measure for the uncertainty of the sample mean growth factor around the population mean, approaches zero with increasing sample size (see Figure 8b) a behavior also known as law of large numbers. For narrow particle number size distributions ( $SD < 0.2 \cdot D_p$ ) the number of particles required to reach better than 1 % precision in growth factor is on the order of 100. The required time to detect this many particles, from now on referred to as minimum sample duration, ranged from about 100 s at 15 nm, to less than 30 s between 35 and 165 nm.

Figure 9 shows average growth factors of ambient aerosol (red circles) and corresponding minimum sample duration (black crosses) measured at constant RH of 85 % on December 3<sup>rd</sup>, 2015. Larger particles ( $D_{p,0} \geq 110 \text{ nm}$ ) were observed to be more hygroscopic ( $\overline{GF} > 1.33$ ). This indicates that larger particles originated from regional background aerosol, which had been processed during transport resulting in a higher soluble fraction (Swietlicki et al., 2008). Below 110 nm particles were found to be nearly hydrophobic ( $\overline{GF} = 1.0 - 1.11$ ). In this size range particles were probably dominated by freshly emitted combustion particles containing soot and water-insoluble organic compounds (Weingartner et al., 1997) as aerosol was sampled near a parking lot.

In addition to the average growth factor, the  $GF$  distribution of the humidified aerosol, its width, and whether it is unimodal or bimodal are examined. Figure 10 compares size distributions of the humidified aerosol obtained by HFIMS at five particle sizes recommended by the EUSAAR project for 20 s (black), and after 200 s (red) of sampling time. All data are for size-selected, initially dry ambient particles humidified to 85 % RH. Note that the short counting times reproduce the final  $GF$

distribution nicely. Widths of the distributions are visually identical. Both short and long sample durations reveal a more hygroscopic mode appearing at 35 nm, growing in prominence at 70 nm, and becoming dominant above 110 nm.

These analyses indicate that the hygroscopic growth factors at the 5 particle sizes could be captured by the HFIMS within 3 minutes, including 15 seconds waiting time to ensure the system reaches steady state following the switching between different  $D_{p,0}$ . Similar measurements using conventional HTDMA systems often take about 30 minutes or more, therefore HFIMS represent an order of magnitude improvement in the measurement speed.

## 5. Conclusions

We present a Humidity-controlled water-based Fast Integrated Mobility Spectrometer (HFIMS) for rapid measurement of particle hygroscopicity.

10 The HFIMS consists of a DMA, an RH-control unit and a Water-based Fast Integrated Mobility Spectrometer (WFIMS) (Pinterich et al., 2017). The WFIMS combines a single high-voltage FIMS (Kulkarni and Wang, 2006a) with the laminar flow water condensation methodologies developed by Hering and coworkers. By detecting particles of different sizes simultaneously, the WFIMS provides rapid mobility based measurements of particle size distributions over a factor of 3 or more in particle diameter, which is sufficient to cover the entire range of growth factor for ambient aerosol particles. Thus, 15 with the combination of DMA, relative humidity control, and WFIMS, the HFIMS can capture the complete growth factor distribution of size selected particles with much improved speed.

Laboratory experiments with NaCl particles showed that HFIMS can reproduce theoretical growth factors within 2 %. The deliquescence transition was observed just over 76 %, in excellent agreement with the theoretical value of 76.5 % (Ming and Russell, 2001). The measured efflorescence relative humidity (43 %) was found to lie within the range of 41 % to 56 % 20 reported in the literature.

The hygroscopicity of ambient aerosols was characterized by keeping sample and sheath RH at 85 % and varying the dry particle size. We found that growth factors of ambient particles ranging from 35 nm to 165 nm could be measured within less than 3 minutes, providing approximately a factor of ten increase in the time resolution. The system will greatly improve our capability to study particle hygroscopic growth, especially for rapidly evolving aerosol populations. ~~or measurements~~ 25 ~~onboard mobile platforms.~~

## Acknowledgments

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## Figures

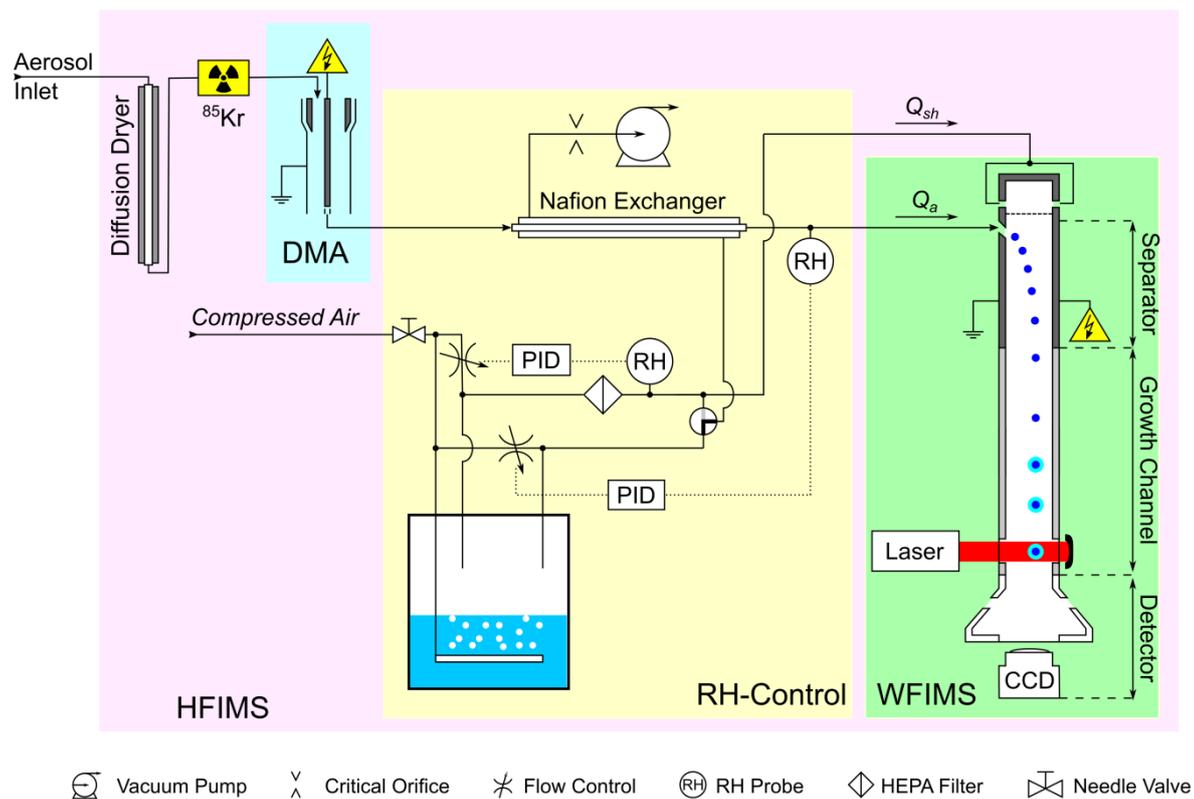
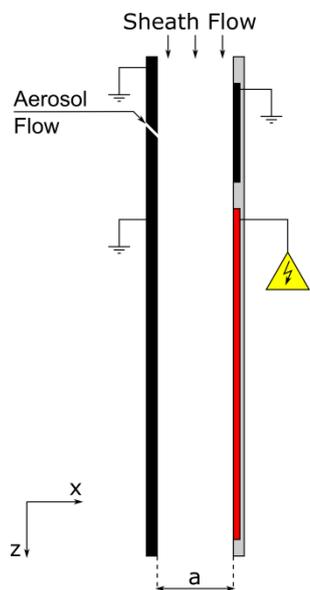
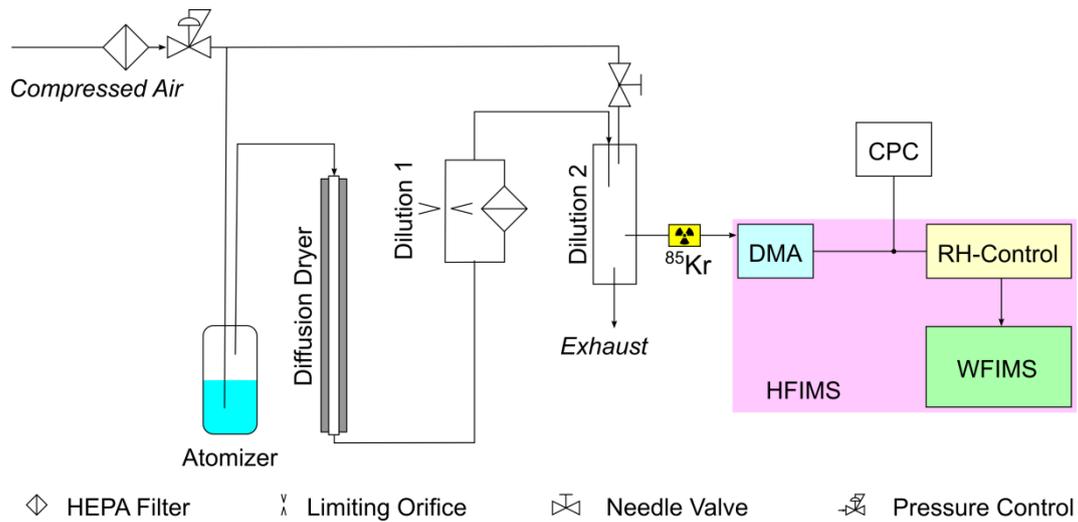


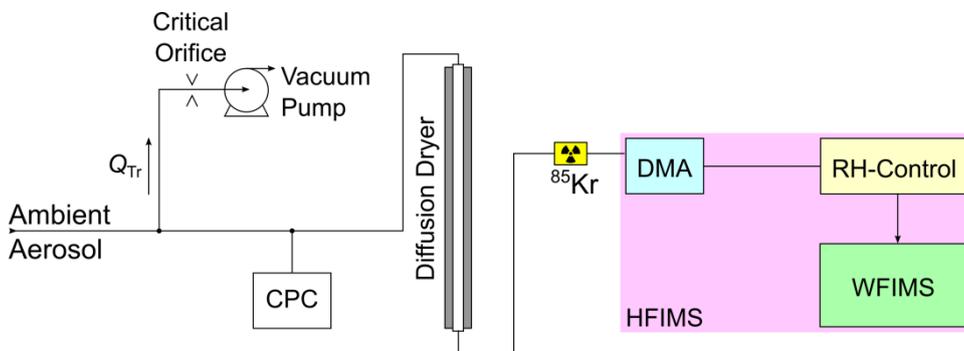
Figure 1. Schematic diagram of the HFIMS.



5 Figure 2 Schematic diagram of the offset electrode used in HFIMS with aerosol inlet on left.



**Figure 3. Experimental setup for laboratory characterization of HFIMS.**



5 **Figure 4 Experimental setup for measuring hygroscopic growth of ambient particles.**

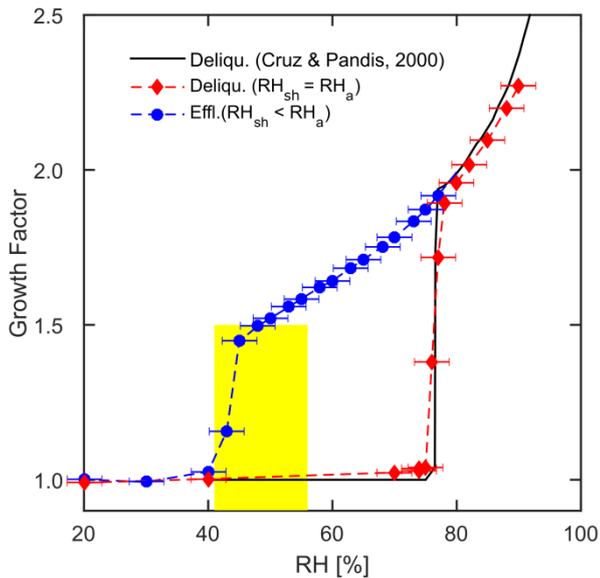


Figure 5. Mean growth factor of NaCl particles ( $D_{p,0} = 50$  nm) measured by the HFIMS as a function of relative humidity (RH) for both deliquescence (red diamonds) and efflorescence (blue circles) branches. Horizontal error bars indicate the RH sensor (Vaisala, Model HMP60) accuracy of  $\pm 3\%$ . Vertical error bars, which represent the standard error of the mean growth factor, are covered by the data point symbols. The solid (black) line represents NaCl ( $D_{p,0} = 100$  nm) deliquescence curve reported by Cruz and Pandis (2000). The yellow area indicates the range of efflorescence transitions reported in literature.

5

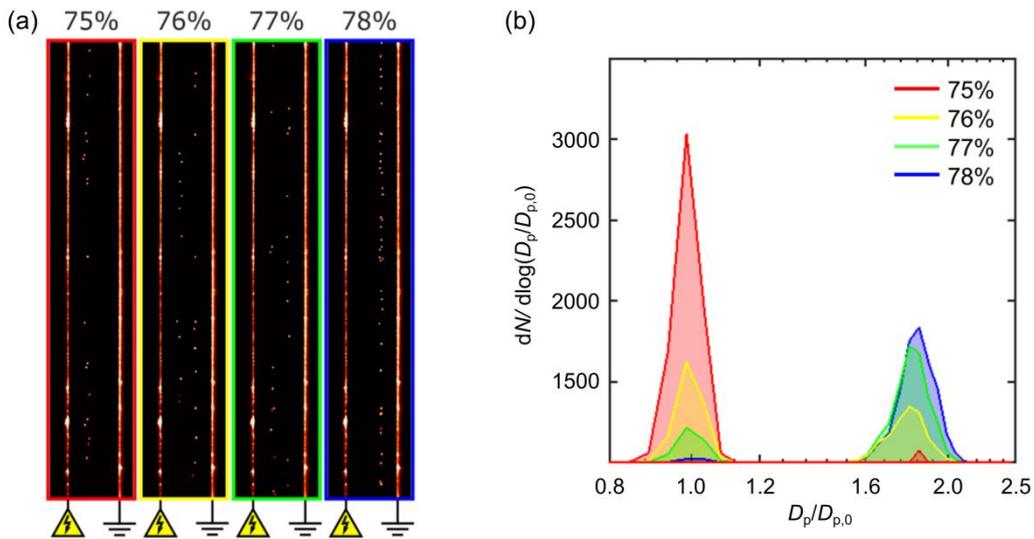


Figure 6 (a) CCD images showing particle positions and (b) number size distributions of NaCl particles ( $D_{p,0} = 50$  nm) measured around the deliquescence point. Red, yellow, green and blue distributions correspond to 75 %, 76 %, 77 % and 78 % RH, respectively.

10

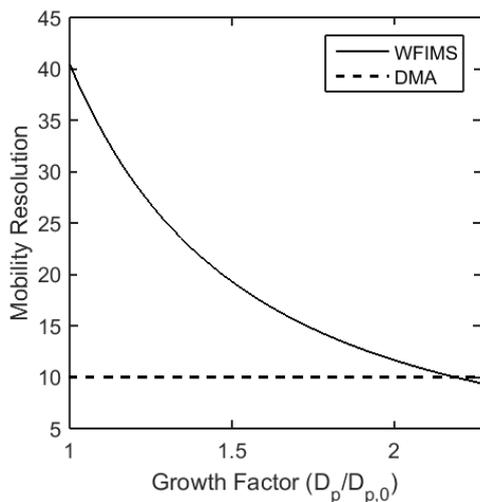


Figure 7 WFIMS **mobility growth factor** resolution (solid line) and typical DMA mobility resolution (dashed line) as a function of growth factor ( $D_p/D_{p,0}$ ) for non-diffusing particles with 50 nm dry size.

5

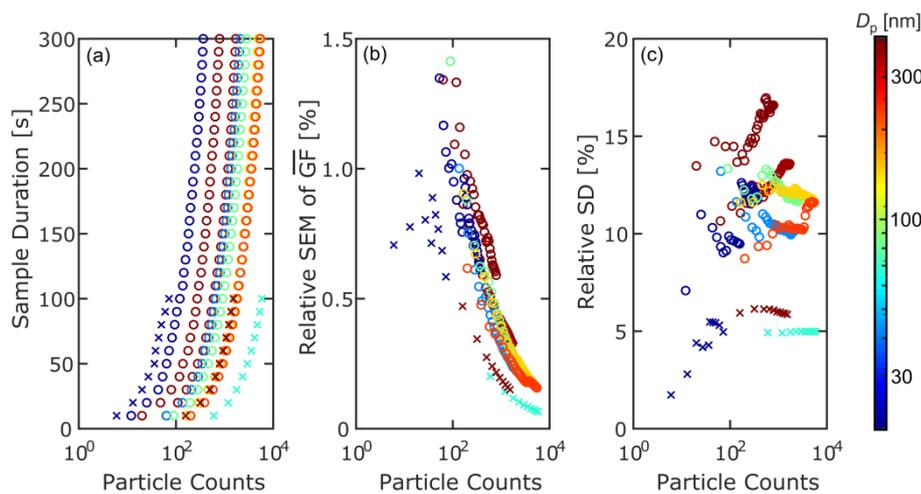


Figure 8 (a) Sample duration, (b) relative standard error (SEM) of the mean particle growth factor  $\overline{GF}$  and (c) relative standard deviation (SD) of the sample particle size distribution as a function of total particle counts for the ambient aerosols sampled on November 6 (crosses) and December 3 (circles) 2015. Color coding represents mean particle wet sizes at RH = 85 %.

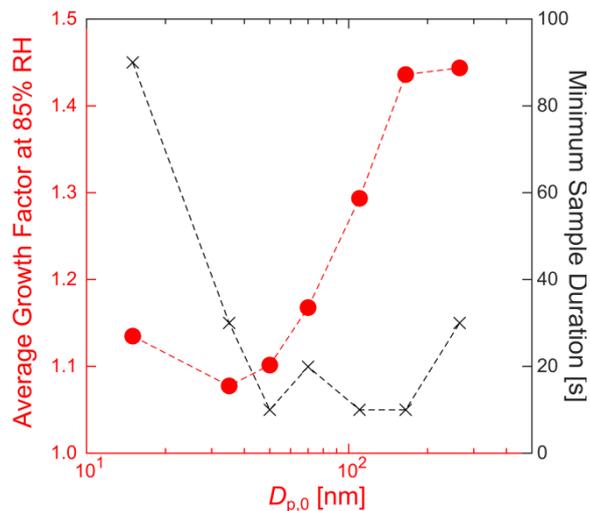
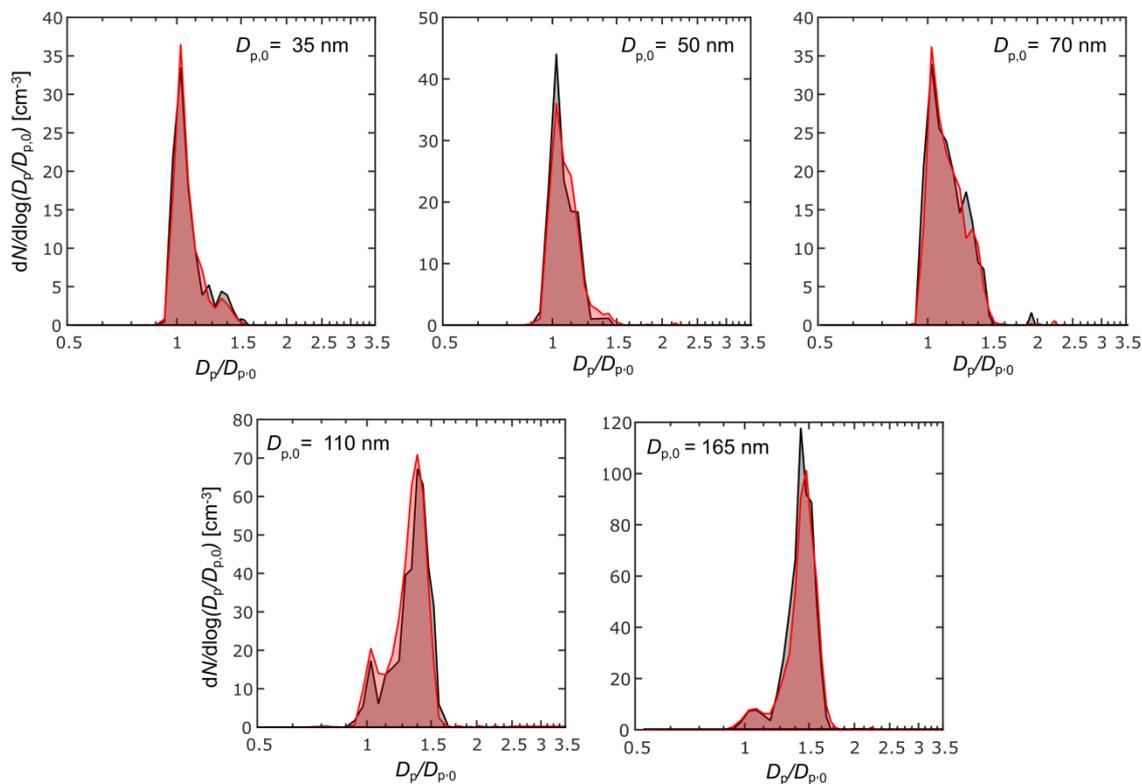


Figure 9. Size dependent average growth factor at RH = 85 % (red circles) and corresponding minimum sample duration (black crosses) of ambient particles sampled on December 3rd, 2015.



5 Figure 10 Comparison of GF distributions of size-selected ambient particles humidified to 85 % for short (20 s, black) and long (200 s, red) sampling periods.

5 Table 1 Dry particle diameter range ( $D_{p,0}^{\min}$  -  $D_{p,0}^{\max}$ ) of four particle hygroscopicity instruments including HFIMS. Diameter ranges of HFIMS, BMI HTDMA and DASH-SP were estimated based on growth factors ranging from 0.8 to 2.2. DASH-SP dry size range was estimated for 5 l/min DMA sheath flow rate, 281 V minimum classifying voltage and 135 nm cut size of the OPC as specified in Sorooshian et al. (2008). The particle dry diameter range of BMI HTDMA was calculated for a typical fieldwork configuration (i. e.  $Q_a = 0.6$  l/min,  $Q_{sh} = 6$  l/min, Lopez-Yglesias et al., 2014) and classifying/scanning voltages ranging from 20 V to 6000 V. The diameter range of H-DMA-APS is listed as specified in Leinert & Wiedensohler (2008).

<u>Instrument</u>	<u><math>D_{p,0}^{\min}</math> [nm]</u>	<u><math>D_{p,0}^{\max}</math> [nm]</u>
<u>HFIMS (This study)</u>	<u>15</u>	<u>205</u>
<u>BMI HTDMA (Lopez-Yglesias et al. 2014)</u>	<u>33</u>	<u>425</u>
<u>DASH-SP (Sorooshian et al. 2008)</u>	<u>170</u>	<u>455</u>
<u>H-DMA-APS (Leinert &amp; Wiedensohler 2008)</u>	<u>800</u>	<u>1600</u>

# Supplemental Information

## S1 WFIMS Configuration

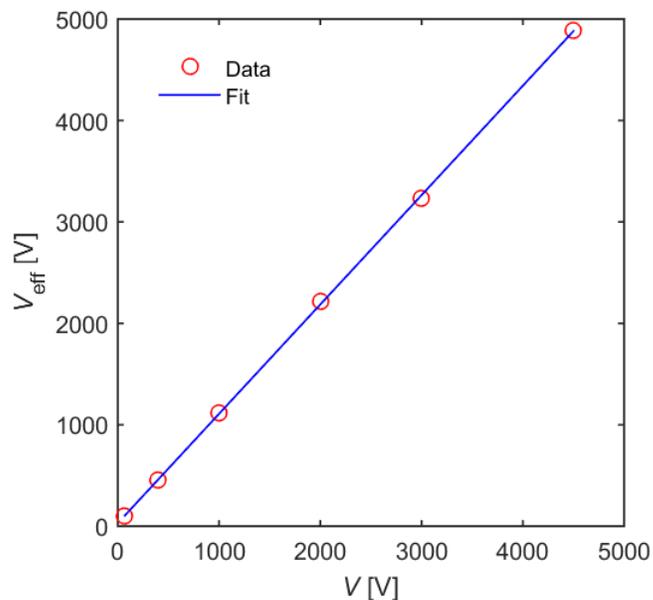
$Q_a$	Aerosol Flow	0.3 l/min
$Q_{sh}$	Sheath Flow	16.2 l/min
$\Delta T_{con}$	Conditioner	-12 °C
$\Delta T_{ini}$	Initiator	+28 °C
$\Delta T_{mod}$	Moderator	-10 °C
$a$	Distance between separator electrodes	0.91 cm
$b$	Width of flow channel = Entrance slit width	12.70 cm
	Entrance slit gap	0.25 mm
$l_s$	Separator length	14.18 cm
$x_{view}$	Range in x-direction of viewing area	0.18 cm–0.73 cm
$y_{view}$	Range in y-direction of viewing area	-3.50 cm– +3.50 cm

5

## S2 HFIMS configuration during laboratory characterization

WFIMS	$V$	Separating Voltage	1000 V	
	$D_{p,min}$	Minimum wet particle diameter	44.8 nm	
	$D_{p,max}$	Maximum wet particle diameter	160.8 nm	
DMA	Type	NDMA		
	$Q_a$	Aerosol Flow	1.3 l/min	
	$Q_{sh}$	Sheath Flow	13 l/min	
	$V$	Classifying Voltage	5332 V	
	$D_{p,0}$	Dry Particle Diameter	50 nm	
RH Control			Deliquescence	Efflorescence
	$RH_a$	Aerosol RH	20 %–90 %	85 %
	$RH_{sh}$	Sheath RH		18.8 %–79.9 %
	RH	Mixed RH		20 %–80 %
HFIMS	$D_{p,min}/D_{p,0}$	Minimum Growth Factor	0.9	
	$D_{p,max}/D_{p,0}$	Maximum Growth Factor	3.2	
CPC	$Q_a$	Aerosol Flow	1 l/min	

### S3 Effective Voltage Calibration



**Figure S1 Calibration curve for the separator voltage of HFIMS.  $V$  and  $V_{\text{eff}}$  are applied and effective voltage, respectively.**

5 To account for non-idealities of the electric field between WFIMS separator electrodes, the applied voltages  $V$  were replaced with effective voltages  $V_{\text{eff}}$  when calculating instrument response mobilities  $Z_p^*$ . The  $V_{\text{eff}}$  values were derived from WFIMS measurements of DMA classified particles at low RH (<10 %). For particles classified with DMA centroid mobilities  $Z_{p,\text{DMA}}$  ranging from  $9.2 \cdot 10^{-7} \text{ m}^2/\text{Vs}$  at  $V=70 \text{ V}$  to  $2.3 \cdot 10^{-8} \text{ m}^2/\text{Vs}$  at  $V=4500 \text{ V}$  (see Sect. S4), the values of  $V_{\text{eff}}$  (see Eq. (2)) were found such that  $Z_p^*$  calculated based on measured particle positions (Eq. (1)) and  $V_{\text{eff}}$  will reproduce  $Z_{p,\text{DMA}}$ . Resulting effective

10 voltages are plotted as a function of applied voltages in Fig. S1 (open red circles). It can be seen that for the entire applied voltage range, i. e. 70–4500 V,  $V_{\text{eff}}$  exceeds  $V$ , and the relationship between  $V_{\text{eff}}$  and  $V$  is linear. Hence following parametrization was used to obtain a general expression for  $V_{\text{eff}}$  based on applied voltages:

$$V_{\text{eff}}(V) = a(V + b). \quad (\text{S1})$$

By fitting  $V_{\text{eff}}$  derived from measurements parameters  $a$  and  $b$  were determined as 1.1 and 27.8 V, respectively. The 27.8 V

15 represent a constant offset, and might be due to an offset of the WFIMS HV power supply, which has a full scale of 10000 V. The scaling factor  $a$  might be related to voltage dependent edge effects of the electric field.

#### S4 HFIMS Configuration during ambient tests

WFIMS	Separating Voltage [V]	Minimum wet particle diameter ( $D_{p,min}$ ) [nm]	Maximum wet particle diameter ( $D_{p,max}$ ) [nm]
	70	12.8	40.4
	400	28.1	94.1
	1000	44.8	160.8
	2000	64.9	253.3
	3000	81.3	338.3
	4500	102.5	461.4
	4500	102.5	461.4

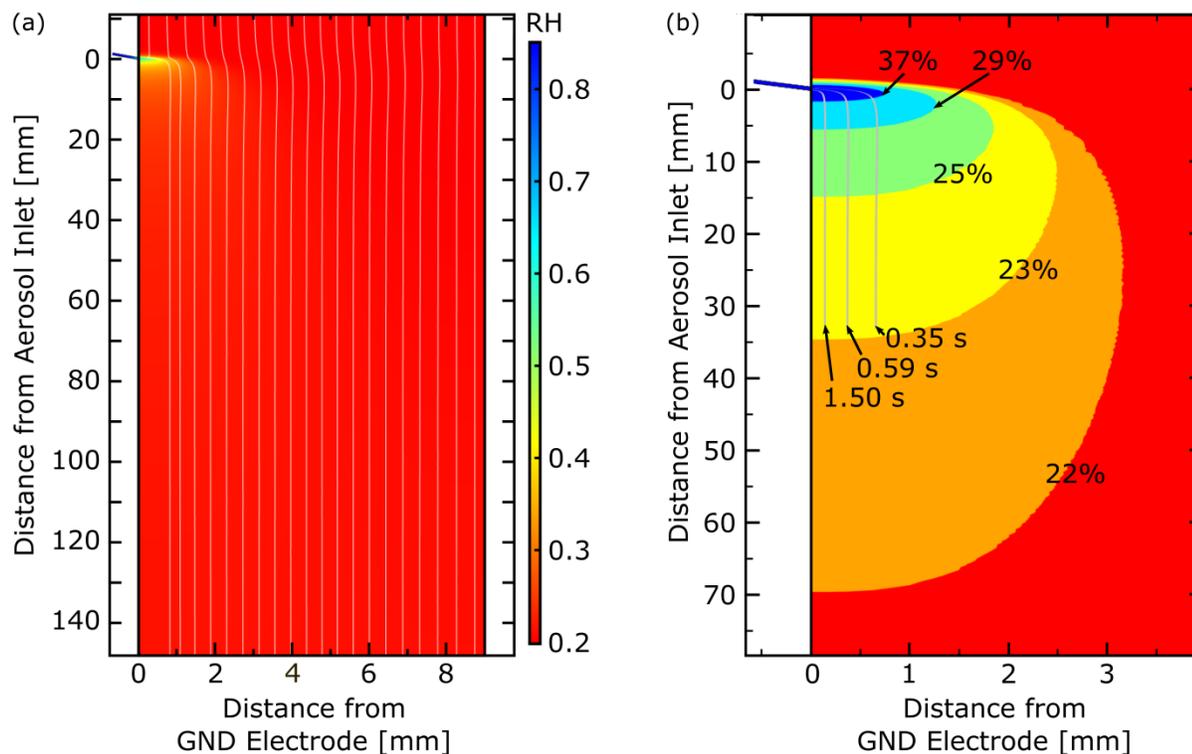
DMA	$Q_a$	Aerosol Flow	0.3 l/min
	$Q_{sh}$	Sheath Flow	3 l/min
	Type	Classifying Voltage [V]	Dry Particle Diameter [nm]
	NDMA	115	15
	NDMA	599	35
	NDMA	1181	50
	NDMA	2202	70
	NDMA	4923	110
	LDMA	1148	165
	LDMA	2355	265

RH Control	$RH_a$	Aerosol RH	85%
	$RH_{sh}$	Sheath RH	
	RH	Mixed RH	

HFIMS	$D_{p,0}$ [nm]	Minimum Growth Factor ( $D_{p,min}/D_{p,0}$ )	Maximum Growth Factor ( $D_{p,max}/D_{p,0}$ )
	15	0.9	2.7
	35	0.8	2.7
	50	0.9	3.2
	70	0.9	3.6
	110	0.7	3.1
	165	0.6	2.7
	265	0.4	1.7

CPC	$Q_a$	Aerosol Flow	1.0 l/min
	$Q_{Tr}$	Transport Flow	5.4 l/min

## S5 Conditioning of aerosol sample RH by the WFIMS sheath flow



**Figure S2 (a) Offset electrode used in HFIMS, with aerosol inlet on left. (b) Enlargement of inlet region showing relative humidity profiles when entering aerosol is at 85 % and sheath flow at 20 %.**

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The single high voltage electrode was configured with an offset such that the high voltage region begins slightly downstream (32 mm) of the introduction of aerosol into the mobility separator. This allows the sheath flow to provide the final humidity conditioning of the aerosol flow prior to mobility classification as the aerosol flow is just 2 % of the total flow. The electrode offset is illustrated in Fig. S2a, where the dimension along the width of the channel is shown enlarged relative to the length. Fig.

10 S2b shows modeled relative humidity profiles at the aerosol inlet obtained using COMSOL Multiphysics®, a numerical modeling package. For instance when the entering aerosol is at 85% RH (0.3 l/min) and the entering sheath flow is at 20 % (16.2 l/min) then at the 32 mm position downstream where the mobility separation begins, the aerosol has reached 23 % RH, within 1.8 % of the desired RH  $21.2 \% = (0.3 \cdot 85 \% + 16.2 \cdot 20 \%) / (0.3 + 16.2)$  after a complete mixing of aerosol and sheath flows. The equilibration time for the aerosol ranges from 0.35 to 1.50 s, as shown Fig. S2b. This approach allows the final RH adjustment of

15 the aerosol to be made within the WFIMS system.